

FORM PTO-1390
(REV. 11-2000)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

24448-0030

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)

09/937464

INTERNATIONAL APPLICATION NO.
PCT/EP00/03020

INTERNATIONAL FILING DATE
April 5, 2000

PRIORITY DATE CLAIMED
April 6, 1999

TITLE OF INVENTION

Silane-based coating mass with a catalytic, oxidative and deodorizing effect

APPLICANT(S) FOR DO/EO/US

Thomas Benthien et al.

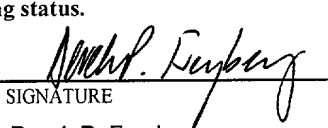
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☐ is attached hereto (required only if not communicated by the International Bureau).
 - b. ☒ has been communicated by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
 - a. ☒ is attached hereto.
 - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ have been communicated by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11 to 20 below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A **FIRST** preliminary amendment.
14. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
15. ☐ A substitute specification.
16. ☒ A change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
18. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☒ Other items or information:

Express Mail Label No. EL912435168US; September 24, 2001

U.S. APPLICATION NO. (if known) 09/937464 INTERNATIONAL APPLICATION NO. PCT/EP00/03020		ATTORNEY'S DOCKET NUMBER 24448-0030		
21. <input checked="" type="checkbox"/> The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)): Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO. \$1000.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$860.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$710.00 International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$690.00 International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00 ENTER APPROPRIATE BASIC FEE AMOUNT =			CALCULATIONS PTO USE ONLY	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).			\$ 860	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$
Total claims	- 20 =		x \$18.00	\$
Independent claims	- 3 =		x \$80.00	\$
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$270.00	\$
TOTAL OF ABOVE CALCULATIONS =				\$ 860
<input checked="" type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.				\$ 430
SUBTOTAL =				\$ 430
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$
TOTAL NATIONAL FEE =				\$ 430
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +				\$ 40
TOTAL FEES ENCLOSED =				\$ 470
				Amount to be refunded: \$
				charged: \$
a. <input checked="" type="checkbox"/> A check in the amount of \$ <u>470</u> to cover the above fees is enclosed. b. <input type="checkbox"/> Please charge my Deposit Account No. <u>08-1641</u> in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>08-1641</u> . A duplicate copy of this sheet is enclosed. d. <input type="checkbox"/> Fees are to be charged to a credit card. WARNING: Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.				
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.				
SEND ALL CORRESPONDENCE TO				
Customer Number 25213		<div style="text-align: center;">  SIGNATURE Derek P. Freyberg NAME 29,250 REGISTRATION NUMBER </div>		

09/937464

J003 24 SEP 2001

PATENTS

Attorney Docket No. 24448-0030

EXPRESS MAIL LABEL INFORMATION - 37 CFR 1.10

Express Mail Label No.: EL912435168US

Date of Deposit: September 24, 2001

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

Thomas Benthien et al. :

App. No.: (not yet known) : Art Unit: (not yet known)
Int'l App. No. PCT/EP00/03020

Filed: (herewith) : Examiner: (not yet known)
Int'l Filing Date: April 5, 2000

For: Silane-based coating mass with a catalytic, oxidative
and deodorizing effect

Box PCT
Commissioner for Patents
Washington, DC 20231

Sir:

PRELIMINARY AMENDMENT

Please amend the above-identified application, before
examination, as follows:

In the Claims:

Cancel the word "CLAIMS" and Claims 1-12, and insert
therefor a new preamble and new Claims 13-26 to read as follows:

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WE CLAIM:

13. A catalytic composition for deodorizing or oxidizing purposes, the composition comprising a coating of a coating material on a support, prepared by a process comprising the steps of:

(i) applying to the support a coating material comprising:

(1) a polycondensate of:

(A) at least one silane of the formula $R_a-Si-X_{(4-a)}$ where each R, which may be the same or different, is a non-hydrolyzable group; each X, which may be the same or different, is a hydroxy group or a non-hydrolyzable group; and a is an integer of 0 to 3 and is greater than 0 for at least 50 mol% of the silanes; or an oligomer derived therefrom, and

(B) optionally, one or more compounds of glass-forming elements; and

(2) particles of at least one transition metal oxide, the weight ratio of the particles of the at least one transition metal oxide to the polycondensate being between 1:10 and 10:1; and

(ii) thermally treating the applied coating material to form the coating.

14. The catalytic composition of Claim 13 where a is greater than 0 for between 50 mol% and 95 mol% of the silanes.

15. The catalytic composition of Claim 13 where the transition metal oxide is selected from the group consisting of the oxides of La, Ce, Ti, Zr, V, Cr, Mo, W, Mn, Fe, Co, Ni, Cu, Ag, Zn, and mixtures thereof.

16. The catalytic composition of Claim 13 where the particles of at least one transition metal oxide have a diameter between 10 nm and 20 μm .

17. The catalytic composition of Claim 13 where the coating has a thickness between 0.01 μm and 500 μm .

18. The catalytic composition of Claim 13 where the support is composed of metal, metal oxide, glass, glass ceramic, ceramic, or porous material.

19. The catalytic composition of Claim 13 where the thermal treatment of step (ii) occurs at between 200 $^{\circ}\text{C}$ and 700 $^{\circ}\text{C}$.

20. The catalytic composition of Claim 13 where the coating material also comprises inorganic particles.

21. The catalytic composition of Claim 13 where the coating is porous.

22. A process for preparing a catalytic composition for deodorizing or oxidizing purposes, the composition comprising a coating of a coating material on a support, the process comprising the steps of:

(i) applying to the support a coating material comprising:

(1) a polycondensate of:

(A) at least one silane of the formula $\text{R}_a\text{-Si-X}_{(4-a)}$

where each R, which may be the same or different, is a non-hydrolyzable group; each X, which may be the same or different, is a hydroxy group or a non-hydrolyzable group; and a is an integer of 0 to 3 and is greater than 0 for at least 50 mol% of the silanes; or an oligomer derived therefrom, and

- (B) optionally, one or more compounds of glass-forming elements; and
- (2) particles of at least one transition metal oxide, the weight ratio of the particles of the at least one transition metal oxide to the polycondensate being between 1:10 and 10:1; and
- (ii) thermally treating the applied coating material to form the coating.

23. The process of Claim 22 where the step of thermally treating the applied coating material occurs without drying or after drying of the applied coating material.

24. A method of deodorizing odor-containing air, comprising passing the odor-containing air over a catalytic composition of Claim 13.

25. The method of Claim 24 where the catalytic composition is maintained at a temperature between 150 °C and 500 °C.

26. A method of oxidizing carbon or organic components present on the surface of the composition of Claim 13, comprising heating the composition to a temperature between 150 °C and 500 °C.

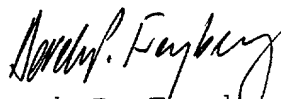
REMARKS

The Amendment

Entry of this amendment is respectfully requested. No new matter is added by the amendment, because the new claims find support in the application as filed. In particular, the new claims remove multiple dependent claims and rewrite the claims in more standard US form.

Claims 13-26 are in this application, Claims 1-12 having been canceled, and Claims 13-26 having been added by this amendment. Entry of the amendment and allowance of the claims are requested.

Respectfully submitted,



Derek P. Freyberg
Attorney for Applicants
Reg. No. 29,250

Customer No.: 25213

Heller Ehrman White & McAuliffe LLP
275 Middlefield Road
Menlo Park, CA 94025-3506
(650) 324-7014
September 24, 2001

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SILANE-BASED COATING MASS WITH A CATALYTIC, OXIDATIVE
AND DEODORIZING EFFECT

5 The invention relates to a catalytic composition, to a
process for preparing it and to the use of the
catalytic composition for the purpose of deodorizing
and oxidizing organic components or carbon.

10 An object of the present invention is to provide
catalytic compositions which are capable of reducing or
eliminating environmental odour pollution (deodorizing)
and which are able to oxidize organic components or
carbon.

15 This objective is surprisingly achieved by means of a
catalytic composition which comprises a coating of a
coating material on a support and is obtainable by
applying the coating material, comprising (1) a
polycondensate of

20 (A) one or more silanes of the general formula (I)



25 in which the radicals R are identical or different
and are non-hydrolysable groups, the radicals X
are identical or different and are hydrolysable
groups or hydroxyl groups and a has the value 0,
1, 2 or 3, with a being greater than 0 for at
least 50 mol% of the silanes, or an oligomer
30 derived therefrom,

(B) if desired, one or more compounds of glass-forming
elements,

and (2) particles of one or more transition metal
oxides, the weight ratio of transition metal oxide
35 particles to polycondensate being from 10:1 to 1:10, to
the support and subjecting the applied coating material
to thermal treatment.

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Specific examples of the functional groups of the radical R^2 are the epoxy, hydroxyl, ether, amino, monoalkylamino, dialkylamino, amide, carboxyl, vinyl, acryloyloxy, methacryloyloxy, cyano, halogen, aldehyde, alkylcarbonyl, and phosphoric acid group. These functional groups are attached to the silicon atom via alkylene, alkenylene or arylene bridging groups, which may be interrupted by oxygen or $-NH-$ groups. The stated bridging groups are derived, for example, from the abovementioned alkyl, alkenyl or aryl radicals. The radicals R^2 contain preferably from 1 to 18, in particular from 1 to 8, carbon atoms.

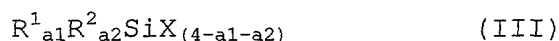
In one preferred embodiment, the silanes (A) comprise a mixture of

(A1) at least one hydrolysable silane of the general formula (II)



in which the radicals X are identical or different and are hydrolysable groups or hydroxyl groups, or an oligomer derived therefrom, and

(A2) at least one organosilane of the general formula (III)



in which R^1 is identical or different at each occurrence and is a non-hydrolysable group, R^2 is identical or different at each occurrence and is a radical which carries a functional group, X has the above definition and a_1 and a_2 have the value 0, 1, 2 or 3, the sum (a_1+a_2) having the value 1, 2 or 3, or an oligomer derived therefrom

in a molar ratio (A1):(A2) of 5-50:50-95.

In the general formula (III), a_1 preferably has the value 1 or 2, a_2 preferably has the value 0, 1 or 2 and the sum (a_1+a_2) preferably has the value 1 or 2.

Particularly preferred hydrolysable silanes (A) and (A1) are tetraalkoxysilanes such as tetraethoxysilane (TEOS). Particularly preferred hydrolysable silanes (A) and (A2) are alkyltrialkoxysilanes, preferably containing $\text{C}_1\text{-C}_8$ alkyl, especially methyltriethoxysilane, aryltrialkoxysilanes, especially phenyltriethoxysilane, dialkyldialkoxysilanes, preferably containing $\text{C}_1\text{-C}_8$ alkyl, especially dimethyldiethoxysilane, and diaryldialkoxysilanes, especially diphenyldiethoxysilane. Silanes containing functional groups (A) and (A2) are, for example, epoxy silanes such as 3-

glycidylxypropyltrimethoxysilane (GPTS) and amino silanes such as 3-aminopropyltriethoxysilane and 3-(aminoethylamino)propyltriethoxysilane (DIAMO).

- 5 In the silane component (A) according to formula (I), a is greater than 0 for at least 50 mol% of the silanes, i.e. at least 50 mol% of the silanes contain at least one non-hydrolysable group R. The silane component (A) preferably comprises from 50 to 95 mol% of silanes
10 having at least one non-hydrolysable group R. With regard to the formulae (II) and (III), the preferred molar ratio of the hydrolysable silane (A1) to the organosilane (A2) in the polycondensate is 5 to 50:50 to 95, preferably from 1:1 to 1:6 and with particular
15 preference from 1:3 to 1:5. A particularly favourable molar ratio is 1:4.

The optional component (B) constitutes glass-forming elements which are preferably dispersible or soluble in
20 the reaction medium. It is possible to use, for example, compounds (halides, alkoxides, carboxylates, chelates, etc.) of lithium, sodium, potassium, rubidium, caesium, beryllium, magnesium, calcium, strontium, barium, boron, aluminium, titanium,
25 zirconium, tin, zinc or vanadium.

To prepare the polycondensate (1), the starting components (A) and, where appropriate, (B) are hydrolysed and condensed. The hydrolysis and
30 condensation are conducted either in the absence of a solvent or, preferably, in an aqueous or aqueous/organic reaction medium, where appropriate in the presence of an acidic or basic condensation catalyst such as HCl, HNO₃ or NH₃. The hydrolysis and
35 condensation preferably take place in the presence of an aqueous acid. The aqueous acids are used preferably in a concentration range of from 0.1 N to 10.0 N. Acids used with preference are hydrochloric, nitric, phosphoric and acetic acid.

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Additionally, during the preparation of the polycondensate, the inorganic particles set out below may be added. During the preparation, preferably, nanoscale inorganic particles, especially in the form of a sol, are added. By way of example, silica sols may act as hydrolytically active compounds in the sol. Suitable for this purpose are commercially customary silica sols, such as the Levasils®, silica sols from Bayer AG, for example.

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When a liquid reaction medium is used, the starting components are soluble in the reaction medium. Particularly suitable organic solvents are water-miscible solvents, such as monohydric or polyhydric aliphatic alcohols, for example, but also aliphatic or aromatic hydrocarbons, such as those having from 5 to 20 carbon atoms, ethers, esters, ketones, amides and alkylamides.

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The hydrolysis and polycondensation preferably take place under the conditions of the sol-gel process, the reaction mixture being used in the viscous sol state to coat the substrate.

20

Where appropriate, the hydrolysis and polycondensation are carried out in the presence of a complexing agent, examples of such agents being nitrates, β -dicarbonyl compounds (e.g. acetylacetonates or acetoacetates), carboxylic acids (e.g. methacrylic acid) or carboxylates (e.g. acetate, citrate or glycolate), betaines, diols, diamines (e.g. DIAMO) or crown ethers.

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The ratio of the hydrolytically active components to the hydrolysable silanes (and, where appropriate, to the glass-forming elements) may be characterized by the value R_{OR} . The R_{OR} value represents the molar ratio of water from the hydrolytically active components (water, aqueous acid, silica sol, etc.) to the abovementioned hydrolysable groups X from the silane components (and,

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In the case of use for oxidation purposes in particular, however, it is possible, besides the particles consisting essentially of transition metal

oxides, to make additional use, in whole or in part, of particles which have the transition metal oxides indicated below at the surface but which otherwise are composed of a different material. In that case the transition metal oxide particles are composed of particles of a material chosen preferably from one of the materials specified below for the inorganic particles, said material being surface-coated with one or more transition metal oxides. Preferably, these particles are coated fully on the surface with the transition metal oxides. For the weight ratio of transition metal oxide particles to polycondensate, these particles are taken into account as a whole as transition metal oxide particles. The particles in question are in particular the particles in the micrometre range, indicated below, which have been provided on the surface, and/or impregnated, with transition metal oxides.

The transition metal oxides in question are, in particular, catalytically active transition metal oxides which have deodorizing and/or oxidizing properties. By transition metals are meant, as is customary, the elements of transition groups I to VIII of the Periodic Table and the lanthanide and actinide elements. With particular preference the transition metal oxide is selected from the oxides of the metals La, Ce, Ti, Zr, V, Cr, Mo, W, Mn, Fe, Co, Ni, Cu, Ag and Zn or from mixtures of these metal oxides.

Preference is given to using transition metal oxide mixtures, with mixtures of the oxides of Mn and Ce with one or two further transition metals, such as mixtures of the oxides of Mn/Co/Ce, Mn/Cu/Ce, Mn/Ni/Ce, Mn/Fe/Ce or Mn/Co/Ni/Ce, being particularly preferred. A further-preferred transition metal oxide mixture is a mixture of the oxides of Cu/V/La. It is also possible to use mixed oxides of the aforementioned transition metals.

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In the transition metal oxide mixtures, the following amounts of the corresponding metal oxides in the metal oxide mixture are preferred: Ce: 1-70% by weight, V: 5-70% by weight, Mn: 20-95% by weight, Fe: 20-95% by weight, Co: 1-50% by weight, Ni: 1-50% by weight, Cu: 1-95% by weight.

Specific examples of transition metal oxides are MnO_2 (pyrolusite), $\gamma\text{-MnO}_2$, Co_3O_4 , Co_2O_3 , CoO and CeO_2 . It is of course also possible to use any other suitable transition metal oxide.

The BET surface area of the particles used is situated, for example, within a range from 1 to $100 \text{ m}^2/\text{g}$.

Besides the transition metal oxide particles, cocatalysts may also be used in the coating material, in amounts for example of from 1 to 5% by weight, based on the transition metal oxide particles. Suitable cocatalysts are, for instance, K, Mg, Ca, Ba and Sr salts and also Al oxide and Sn oxide. Examples of suitable salts are the corresponding halides, hydroxides, nitrates, carbonates, or phosphates. They may be added, for example, by mixing the cocatalyst with the transition metal oxide particles or with the mixtures of the transition metal oxide particles prior to addition to the polycondensate, or by separate addition of the cocatalyst to the coating material. In the former case, it is preferred to use powders, and in the latter case it is preferred to use readily soluble salts of the cocatalyst.

The coating material may also include inorganic particles, which may be added during the preparation of the polycondensate or of the coating material or thereafter. These particles may be nanoscale inorganic particles or particles in the micrometre range. It is also possible to add particles of both orders of magnitude, in which case the particles in the

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micrometre range are employed in particular when the catalytic composition is used for oxidizing organic components or carbon.

- 5 The inorganic particles may be composed of any desired materials, with oxides being preferred. Preferred oxides are oxides of Si and Al (especially boehmite). The particles may be added, for example, in the form of powders or, especially the nanoscale particles, in the
10 form of sols.

The nanoscale inorganic particles preferably possess an average particle size of up to 300 nm, in particular up to 100 nm and with particular preference up to 50 nm.

- 15 The particles may be added in colloidal form. In this case they can comprise sols or dispersible powders. Specific examples of nanoscale inorganic particles are SiO_2 , Al_2O_3 , SnO_2 , iron oxides or carbon (carbon black and graphite), especially SiO_2 . Very particular
20 preference is given to using silica sols as nanoscale inorganic particles.

- Especially if the catalytic compositions are to be used as oxidative compositions, inorganic particles in the
25 micrometre range may also be added to the coating material. They serve to structure the coating and to produce cavities. These particles possess an average particle diameter of, for example, from 1 to 500 μm , preferably from 10 to 300 μm . They are preferably
30 oxide- and/or hydroxyl-containing compounds of the elements from main groups III and IV, such as aluminium oxides or silicon oxides. They may have been activated. Examples that may be mentioned include kieselguhr, alumina 90, silica gel 40 or silica gel 60, produced by
35 the company Merck.

Prior to their use, the abovementioned inorganic particles in the micrometre range may be impregnated with metal salts or mixtures of metal salts, such as

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chlorides, phosphates, formates, nitrates or acetates, and then treated at elevated temperatures in order to generate catalytically active metal oxides on the surface. Preference is given to using metal nitrates or metal acetates, since the anions form volatile products when treated within the temperature range used. Metals used are the transition metals specified for the transition metal oxide particles. In this case particles are obtained which are provided on the surface with transition metal oxides, which are used in accordance with the invention as transition metal oxide particles, and which are taken into account as a whole for the weight ratio of transition metal oxide particles to polycondensate.

The coating material may also comprise further additives. It is possible, for example, to use additives suitable for adjusting viscosity and/or, in particular, for generating cavities during the thermal treatment of the coating materials. For this purpose it is possible, for example, to use customary thickeners. Specific examples are cellulose derivatives, such as hydroxypropylcellulose, starch, modified starch, polyvinyl alcohol and glycols, e.g. polyethylene glycol. Preference is given to using cellulose derivatives, especially hydroxypropylcellulose. In addition it is also possible to use the additives customary in catalytic compositions, such as pigments (e.g. black pigments).

The viscosity of the sol mixed with the transition metal oxide particles may also be adjusted, if desired, by removing or adding a solvent, e.g. one of those mentioned above. In this form, the sol is usually also storable for long periods. Where appropriate, it may be activated by adding water or aqueous acid, in which case the coating material is preferably used within one month.

The coating material is applied to the support by customary coating methods. Applicable techniques are, for example, dipping, flow coating, spin coating, spraying or brushing.

Suitable supports are, for example, those of metals such as stainless steel, steel, copper, brass and aluminium; metal oxides, glasses such as float glass, borosilicate glass, lead crystal or silica glass; glass ceramics, and ceramics such as Al_2O_3 , ZrO_2 , SiO_2 mixed oxides, or else enamel, but also porous supports such as porous ceramics, for example. The shape of the supports is arbitrary. The supports in question may be planar or structured. Particularly suitable supports are those in the form of meshes, honeycombs or nets, such as wire meshes, examples being steel wire meshes, ceramic honeycombs or wire nets.

The supports may be pretreated prior to the application of the coating material. For example, they are subjected to cleaning, using commercially customary alkaline cleaners, for example. It is likewise possible, for example, by heat-treating steel supports and forming chromium oxide whiskers on the surface, to bring about substantially improved adhesion of the coating material to steel supports.

The resulting coating is subjected to initial drying, if desired, and then heat-treated. This can be done at temperatures of from 200°C to 700°C , preferably from 300°C to 400°C . The heat treatment may be carried out in air or in an inert gas such as nitrogen or argon. The heat treatment may also take place if desired by means of IR or laser radiation. The heat treatment may be accompanied, for example, by drying, curing or consolidation or compaction of the coating material.

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The coating operation is preferably performed so as to give coat thicknesses of from 0.01 to 500 μm , preferably from 1 to 500 μm . Where the catalytic compositions are used for the purpose of deodorizing, coat thicknesses of from 30 to 100 μm , in particular from 25 to 75 μm , are preferred. Where the catalytic compositions are used as oxidatively active surfaces, coat thicknesses of from 1 to 10 μm are suitable when using transition metal oxides having an average particle size of less than 200 nm. The catalytic compositions which serve as oxidatively active surfaces, and which additionally comprise inorganic particles in the micrometre range, preferably have coat thicknesses of from 100 to 400 μm .

The catalytic compositions of the invention may have a porous or a non-porous coating. Preferably, the catalytic compositions have porous coatings. The pores may comprise microscopically visible cavities on the surface and/or relatively fine micropores. The cavities visible on the surface under the microscope have an approximately globular morphology (hemispheres) and their diameter is from about 1 to 5 μm . Their extent and form in the interior of the coat cannot be determined by microscopy. The determination of the BET surface areas of preferred embodiments indicates that relatively fine micropores are present therein alternatively or additionally.

The catalytic composition of the invention has a deodorizing effect; in other words, odour pollution caused by substances can be reduced or avoided completely. The deodorizing activity is found in particular at temperatures above 150°C, for example at temperatures from 150 to 500°C, preferably from 200 to 350°C. The odour-polluted air is guided past the catalytic composition at elevated temperatures. As it passes, substances present in the air are degraded.

5 The catalytic composition is also capable of oxidizing organic components or carbon, such as carbon black or graphite, which are present, for example, on the surface of the catalytic composition. The oxidizing activity is found in particular at the temperature ranges indicated above.

10 On the basis of these properties, the catalytic composition is preferably employed where odour pollution events may occur or where particularly "odour-neutral" air (that is, air with as little of additional substances as possible) is desirable or where the oxidation of organic components or carbon is
15 desired. In general, the catalytic compositions may be used, for example, in livestock farming, food processing, for example in fish processing or in cheese dairies, in fabrication processes, in waste processing, or generally in chemical industry plants, but also in
20 living spaces. Specific examples of fields of use are therefore toilets of all kinds, baths, means of transport, such as cars, combustion units, especially their exhaust gas installations, e.g. the (diesel) exhaust of a car, caravans, tanks, animal stalls,
25 petrol stations, digestion towers of sewage treatment plants, composting units, manure stations, silos, waste air units of all kinds, gas masks, wardrobes, nappies, refuse containers or gas sensors. The catalytic compositions are preferably used in these fields in
30 such a way that they are located directly on any surface of the article in question, in which case this surface acts as the substrate, or they are located in an additional facility, connected where necessary via a connecting line, within or in the vicinity of the
35 article or of the space within which the articles are located.

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EXAMPLES

A. Preparation of the silane sols

5

Silane sol 1: MTKS sol, $R_{OR} = 0.4$

A mixture of 1069.86 g (6.0 mol) of methyltriethoxysilane and 312.48 g (1.5 mol) of tetraethoxysilane is divided into two portions (portion 1 and portion 2) of equal weight.

To portion 1, 246.84 g of silica sol (Levasil 300/30, aqueous, basic, Bayer AG) are added with thorough stirring. After an emulsion has formed (about 30 s), 5.60 g of 36% strength by weight HCl are added. After brief stirring (30-50 s) the reaction mixture becomes clear with heating. Portion 2 is added quickly and all at once to this reaction mixture. After a short time, the reaction mixture becomes cloudy owing to a colourless precipitate (NaCl). This is followed by stirring with cooling in an ice bath for 15 minutes. The silane hydrolysate is left to stand at room temperature for 12 h and decanted from the sedimented solid, thus giving the ready-to-use MTKS sol.

25 Silane sol 2: MDKS sol, $R_{OR} = 0.2$

35.10 g of silica sol (Levasil 300/30, aqueous, basic, Bayer AG) and 1.10 g of 36% strength by weight HCl are added simultaneously to a mixture of 356.62 g (2.0 mol) of methyltriethoxysilane and 74.14 g (0.5 mol) of dimethyldiethoxysilane, with thorough stirring. After brief stirring (30-50 s) the reaction mixture becomes clear with heating. After a short time, the reaction mixture becomes cloudy owing to a colourless precipitate (NaCl). This is followed by stirring with cooling in an ice bath for 15 minutes. The silane hydrolysate is left to stand at room temperature for 12 h and decanted from the sedimented solid, thereby giving the ready-to-use MDKS sol.

Silane sol 3: MPTKS sol, ROR = 0.4

3.29 g of silica sol (Levasil 300/30, aqueous, basic, Bayer AG) and 0.13 g of 36% strength by weight HCl are added simultaneously to a mixture of 11.59 g (0.065 mol) of methyltriethoxysilane, 3.61 g (0.015 mol) of phenyltriethoxysilane and 4.17 g (0.020 mol) of tetraethoxysilane, with thorough stirring. After brief stirring (30-50 s) the reaction mixture becomes clear with heating. After a short time, the reaction mixture becomes cloudy owing to a colourless precipitate (NaCl). This is followed by stirring with cooling in an ice bath for 15 minutes. The silane hydrolysate is left to stand at room temperature for 12 h and decanted from the sedimented solid, thereby giving the ready-to-use MPTKS sol.

Silane sol 4: MPrTKS sol, ROR = 0.4

7.00 g of silica sol (Levasil 300/30, aqueous, basic, Bayer AG) and 0.23 g of 32% strength by weight HCl are added simultaneously to a mixture of 15.00 g (0.084 mol) of methyltriethoxysilane, 14.95 g (0.091 mol) of n-propyltrimethoxysilane and 8.96 g (0.043 mol) of tetraethoxysilane, with thorough stirring. After brief stirring (30-50 s) the reaction mixture becomes clear with heating. After a short time, the reaction mixture becomes cloudy owing to a colourless precipitate (NaCl). This is followed by stirring with cooling in an ice bath for 15 minutes. The silane hydrolysate is left to stand at room temperature for 12 h and decanted from the sedimented solid, thereby giving the ready-to-use MPrTKS sol.

Silane sol 5: MD sol, ROR = 0.4

5.04 g of 0.1 N HCl are added to a mixture of 35.66 g (0.2 mol) of methyltriethoxysilane and 7.41 g (0.05 mol) of dimethyldiethoxysilane, with thorough stirring. After brief stirring (30-50 s) the reaction mixture becomes clear with heating. The silane

hydrolysate is left to stand at room temperature for 12 h, thereby giving the ready-to-use MD sol.

B. Preparation of the catalyst mixtures

- 5 The catalyst mixtures used are mixtures of commercial transition metal oxide powders from Ferro or Aldrich:

MnO₂: Powder from Ferro, predominantly MnO₂
(pyrolusite), with a little γ-MnO₂ and a little
10 MnO₂
Co_yO_x: Powder from Ferro, predominantly Co₃O₄, with a
very little CoO

Catalyst mixture 1: Mn/Co/Ce

- 15 Catalyst mixture 1 is prepared by intimately mixing 800.00 g of MnO₂, 100.00 g of Co_yO_x and 100.00 g of CeO₂.

Catalyst mixture 2: Mn/Co/Ce

- 20 Catalyst mixture 2 is prepared by intimately mixing 800.00 g of MnO₂, 150.00 g of Co_yO_x and 50.00 g of CeO₂.

Catalyst mixture 3: Mn/Cu/Ce

- 25 Catalyst mixture 3 is prepared by intimately mixing 650.00 g of MnO₂, 300.00 g of Cu₂O and 50.00 g of CeO₂.

Catalyst mixture 4: Mn/Co/Ni/Ce

- 30 Catalyst mixture 4 is prepared by intimately mixing 700.00 g of MnO₂, 100.00 g of Co_yO_x, 150.00 g of NiO and 50.00 g of CeO₂.

C. Preparation of the coating materials

Example 1

- 35 1000.00 g of catalyst mixture 1 are stirred at room temperature for 2 h with 300.00 g of silane sol 1 and 233.33 g of ethanol. Then 32.35 g of 10% strength by weight aqueous hydrochloric acid are added for activation (increasing the R_{OR} value from 0.4 to 0.8),

the mixture is stirred at room temperature for at least 2 h, and the ready-to-use coating suspension is obtained.

5 **Example 2**

1000.00 g of catalyst mixture 2 are stirred at room temperature for 2 h with 200.00 g of silane sol 1 and 350.00 g of ethanol. Then 23.49 g of 10% strength by weight aqueous hydrochloric acid are added for
10 activation (increasing the R_{OR} value from 0.4 to 0.8), the mixture is stirred at room temperature for at least 2 h, and the ready-to-use coating suspension is obtained.

15 **Example 3**

1000.00 g of catalyst mixture 3 are stirred at room temperature for 1 h with 400.00 g of silane sol 2 and 185.00 g of ethanol. Then 47.97 g of 10% strength by weight aqueous hydrochloric acid are added for
20 activation (increasing the R_{OR} value from 0.2 to 0.6), the mixture is stirred at room temperature for at least 4 h, and the ready-to-use coating suspension is obtained.

25 **Example 4**

1000.00 g of catalyst mixture 3 are stirred at room temperature for 1 h with 18.00 g of silane sol 3 and 25.00 g of ethanol. Then 1.52 g of 10% strength by weight aqueous hydrochloric acid are added for
30 activation (increasing the R_{OR} value from 0.4 to 0.7), the mixture is stirred at room temperature for at least 2 h, and the ready-to-use coating suspension is obtained.

35 **Example 5**

1000.00 g of catalyst mixture 4 are stirred at room temperature for 1 h with 40.00 g of silane sol 5 and 11.00 g of ethanol. Then 4.66 g of 10% strength by weight aqueous hydrochloric acid are added for

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activation (increasing the R_{OR} value from 0.4 to 0.8), the mixture is stirred at room temperature for at least 2 h, and the ready-to-use coating suspension is obtained.

5

D. Coating and heat treatment (especially for deodorizing purposes)

The support material used is steel wire mesh (diameter about 5 cm, height about 1 cm) or ceramic honeycombs.

10 The steel meshes are first of all degreased using a commercial alkaline cleaner and then rinsed thoroughly with deionized water, before being dried at room temperature. The dry steel meshes are subsequently treated at 500°C for 1 h.

15

Coating takes place by impregnating the steel wire meshes or the ceramic honeycombs in one of the coating materials (coating suspensions) described in section C. The excess coating suspension is removed by blowing with compressed air. After drying at room temperature (2 h), the coating is solidified by heat treatment. For this purpose the coated supports are heated from room temperature to 300-400°C over the course of 1 h, held at 300-400°C for 1 h, and then cooled to room temperature over 6 h.

20

25

Alternatively, the heat treatment may also be effected by direct placement of the dried, coated supports into an oven preheated to 300-400°C and rapid cooling of the hot supports to room temperature over a few minutes.

30

The thicknesses of the thermally solidified coats are typically in the range 25-75 μm . The coat thicknesses may be set, for example, on the one hand by way of the viscosity of the coating suspension (which can be adjusted, for example, by adding an appropriate amount of ethanol), on the other by way of the pressure of the compressed air or the time of action of the compressed air during removal of the excess coating suspension.

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E. Catalytic composition 1 (especially for oxidizing)

E.1 Preparation of an Mn/Cu/Ce catalyst on alumina particles

40.47 g (0.141 mol) of $\text{Mn}(\text{NO}_2)_2 \cdot 6 \text{H}_2\text{O}$, 11.63 g
5 (0.050 mol) of $\text{Cu}(\text{NO}_3)_2 \cdot 3 \text{H}_2\text{O}$ and 15.20 g (0.035 mol)
of $\text{Ce}(\text{NO}_3)_3 \cdot 6 \text{H}_2\text{O}$ are dissolved in a mixture of
30.00 g of ethanol and 30.00 g of water at 50°C. 100.00
g of alumina 90 (active, acidic (alternatively, neutral
or basic can also be used), particle size 63-200 μm ,
10 from Merck) are added to this solution and the solvent
mixture is evaporated off with stirring at 50-70°C for
3 h. The alumina impregnated with the metal nitrates is
subsequently treated at 500°C for 1 h. Analogously, it
is also possible to use the corresponding molar amounts
15 of metal acetates or, instead of aluminium 90, the
further Merck products silica gel 40, particle size 63-
200 μm , silica gel 60, particle size 63-200 μm , or
kieselguhr, particles size approximately 100 μm .

E.2 Coating material

150.00 g of the above-described Mn/Cu/Ce catalyst (E.1)
on the alumina particles are intimately mixed with
50.00 g of catalyst mixture 1. 100.00 g of a 5%
strength by weight solution of hydroxypropylcellulose
25 in ethanol are added with stirring to this mixture.
140.00 g of silane sol 2 are activated (increasing the
 R_{OR} value from 0.2 to 0.8) by addition of 22.67 g
(1.26 mol) of water, with stirring, and the mixture is
stirred at room temperature for 30 minutes. The
30 activated MDKS sol is added to the above-described
mixture of Mn/Cu/Ce catalyst, catalyst mixture 1 and
hydroxypropylcellulose in ethanol, at room temperature
with stirring, and the mixture is then stirred at room
temperature for 15 minutes to give the ready-to-use
35 coating material.

E.3 Coating and thermal solidification

The support material used is steel substrates (metal
panels 10 x 10 cm). The steel substrates are first of

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all degreased using a commercial alkaline cleaner, then rinsed thoroughly with deionized water, and subsequently dried at room temperature. The dry steel substrates may then be treated at 500°C for 1 h.

5

The cleaned, or cleaned and heat-treated, steel substrates are flooded with the coating material. The coated steel substrates are dried at room temperature for 1 h, then heated from room temperature to 500°C over 1 h, held at 500°C for 1 h, and then cooled to room temperature over 6 h.

The thicknesses of the thermally solidified coats are typically in the range 150-400 μm , depending on the support material used and the amount of coating material used.

F. Catalytic composition 2 (particularly for oxidizing)

F.1 Preparation of a mixed oxide catalyst by coprecipitation of Mn/Co/Ce

121.42 g (0.423 mol) of $\text{Mn}(\text{NO}_3)_2 \cdot 6 \text{H}_2\text{O}$, 14.55 g (0.050 mol) of $\text{Co}(\text{NO}_3)_2 \cdot 6 \text{H}_2\text{O}$ and 9.42 g (0.022 mol) of $\text{Ce}(\text{NO}_3)_3 \cdot 6 \text{H}_2\text{O}$ are dissolved in 350.00 g of water at 80-90°C. The precipitating reagent used is a solution of 66.77 g (1.19 mol) of KOH in 300.00 g of water to which 1.60 g of Tween 80 (polyoxyethylene(20)sorbitan monooleate from Aldrich) and 1.60 g (0.012 mol) of 1-octanol are added with stirring. The above-described solution of the metal nitrates is admixed with the above-described precipitating reagent over 5 minutes at 80-90°C with stirring. The homogeneous, loam-coloured suspension thus obtained is stirred at 90°C for 2 h more and then filtered to remove the precipitate, which is washed twice with 150 g of water each time and once with 50 ml of ethanol. The precipitate is initially dried at 70°C for 1 h and then heated from room temperature to 600°C

over 1 h, held at 600°C for 2 h, and then cooled to room temperature over 2 h.

5 The greenish powder obtained in this way is suspended
in 250.00 g of water at 90°C and then, following the
addition of 1.60 g of Tween 80 and 1.60 g (0.012 mol)
of 1-octanol, it is redispersed over the course of 30
minutes using an ultrasonic disintegrater. It is then
10 filtered to remove the precipitate, which is washed
twice with 50 g of water each time and once with 100 g
of ethanol. The precipitate is initially dried at 70°C
for 1 h and then heated from room temperature to 250°C
over 1 h, held at 250°C for 1 h, and then cooled to
room temperature over 2 h. This gives a fine, greenish
15 catalyst powder.

F.2 Coating material

3.00 g of silane sol 2 are activated (increasing the R_{OR}
value from 0.2 to 0.8) by addition of 0.48 g
20 (0.027 mol) of water with stirring, and the mixture is
stirred at room temperature for 30 minutes. 10.00 g of
the catalyst powder described in section F.1 and 8.00 g
of ethanol are added to the activated MDKS sol with
stirring, and the mixture is stirred at room
25 temperature for 30 minutes to give the ready-to-use
coating material.

F.3 Coating and thermal solidification

The support material used is steel substrates (metal
30 panels 10 × 10 cm). The steel substrates are first of
all degreased using a commercial alkaline cleaner, then
rinsed thoroughly with deionized water, and
subsequently dried at room temperature. The dry steel
meshes may then be treated at 500°C for 1 h. The
35 cleaned, or cleaned and heat-treated, steel substrates
are flooded with the coating material. The coated steel
substrates are dried at room temperature for 1 h, then
heated from room temperature to 300-600°C over 1 h,

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held at 300-600°C for 1 h, and then cooled to room temperature over 6 h.

- 5 The thicknesses of the thermally solidified coats are typically in the range 1-10 μm , depending on the amount of coating material used.

G. Evaluation

10 **Method of determining the deodorizing activity**

About 100 mg of the following test substances are introduced into a circulating-air oven preheated to 300°C (catalyst temperature about 300°C, support: steel wire mesh):

- 15 pyrazine, thiazole, maltol, vanillin and 2,4-decadienal.

- 20 The test substances evaporate in the hot oven, with the vapours being passed as off-gases (off-gas flow: 0.5-1.2 l/s) by the stream of circulating air through an outlet port without a catalyst and an outlet port with catalyst to a downstream sample collector. The collected samples are analysed by means of GC-MS spectroscopy. The spectra are used to determine
- 25 breakdown rates for the test substances in the off-gas stream that passes over the catalyst in comparison to the off-gas stream which does not pass over a catalyst (principle: relative measurement on an experimental system). The breakdown rates are indicated below in %.

30

Catalyst	Pyrazine	Thiazole	Maltol	Vanillin	Decadienal
Pd/Pt	0	0	90	90	-
cat(*1)					
(*2)	83	88	73	78	65
(*3)	69	56	74	70	-

(*1): Palladium, metallic, on steel wire nets, commercial catalyst

(*2): Inventive Mn/Co/Ce-MTKS sol cat., coating material of Example 1

(*3): Inventive Mn/Cu/Ce-MDKS sol cat., coating material of Example 3.

5

It is found that the catalytic compositions of the invention are capable of breaking down not only the other test substances but also heterocycles such as pyrazine and thiazole. This is not possible with commercially customary palladium catalysts. Also, with the catalytic compositions of the invention, there is no loss of catalytic activity after ten test cycles. In contrast, the commercially customary palladium catalyst is poisoned by heterocycles such as thiazole, losing catalytic activity with time.

Evaluation of the oxidizing capacity

[Test method according to DIN 51 171, "Testing of the self-cleaning capacity of continuously self-cleaning enamel coatings"]

Defined amounts (in each case 20-25 mg) of soya oil or engine oil are applied dropwise to the samples under investigation, at five points located on a circle, and after each dropwise addition are burnt by a one-hour heat treatment at 250°C, until a visible lustre appears as a result of the accumulation of unburned residues. The number of cycles until lustre occurs is used for the assessment.

30

Coating	Oil	Number of cycles to lustre
(*1)	Soya	4-5
(*2)	Soya	15-17
(*3)	Soya	13-15
(*4)	Engine	10-12

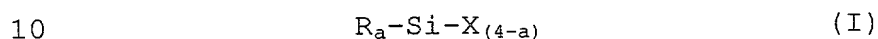
(*1): Commercially customary, oxidative enamel, containing Fe/Mn oxides

- 10

The catalytic compositions of the invention (coat thicknesses between 150-400 micrometres) possess high absorbency, owing to the cavities which exist in the coating, and hence have a good spreading capacity for oils. In contrast, the glass-like enamels have a low absorbency and spreading capacity.

CLAIMS

1. Catalytic composition for deodorizing or oxidizing purposes which comprises a coating of a coating material on a support and is obtainable by applying the coating material, comprising (1) a polycondensate of (A) one or more silanes of the general formula (I)



in which the radicals R are identical or different and are non-hydrolysable groups, the radicals X are identical or different and are hydrolysable groups or hydroxyl groups and a has the value 0, 1, 2 or 3, with a being greater than 0 for at least 50 mol% of the silanes, or an oligomer derived therefrom,

(B) if desired, one or more compounds of glass-forming elements,

and (2) particles of one or more transition metal oxides, the weight ratio of transition metal oxide particles to polycondensate being from 10:1 to 1:10, to the support and subjecting the applied coating material to thermal treatment.

2. Catalytic composition according to Claim 1, characterized in that a is greater than 0 for from 50 to 95 mol% of the silanes.

3. Catalytic composition according to Claim 1 or 2, characterized in that the transition metal oxide is selected from the oxides of the metals La, Ce, Ti, Zr, V, Cr, Mo, W, Mn, Fe, Co, Ni, Cu, Ag and Zn or mixtures thereof.

4. Catalytic composition according to one of Claims 1 to 3, characterized in that the particle diameter of

the transition metal oxide particles is from 10 nm to 20 μm .

5. Catalytic composition according to one of Claims 1 to 4, characterized in that the thickness of the coating is from 0.01 to 500 μm .

6. Catalytic composition according to one of Claims 1 to 5, characterized in that the support is composed of metal, metal oxide, glass, glass ceramic, ceramic or porous material.

7. Catalytic composition according to one of Claims 1 to 6, characterized in that the coating material, immediately or after drying, has been treated at an air temperature range from 200 to 700°C.

8. Catalytic composition according to one of Claims 1 to 7, characterized in that the coating material additionally includes inorganic particles.

9. Catalytic composition according to one of Claims 1 to 8, characterized in that the coating formed from the coating material is porous.

10. Process for preparing a catalytic composition for deodorizing or oxidizing purposes which comprises a coating of a coating material on a support, wherein (1) a polycondensate of

(A) one or more silanes of the general formula (I)



in which the radicals R are identical or different and are non-hydrolysable groups, the radicals X are identical or different and are hydrolysable groups or hydroxyl groups and a has the value 0, 1, 2 or 3, with a being greater than 0 for at

least 50 mol% of the silanes, or an oligomer derived therefrom, and

(B) if desired, one or more compounds of glass-forming elements

5 is mixed with (2) particles of one or more transition metal oxides in a weight ratio of transition metal oxide particles to polycondensate of from 10:1 to 1:10, a coating material comprising this mixture is applied to the support and, immediately or after drying, is
10 subjected to heat treatment.

11. Use of the catalytic composition according to one of Claims 1 to 9 for deodorizing.

15 12. Use of the catalytic composition according to one of Claims 1 to 9 for oxidizing organic components or carbon.

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ABSTRACT

SILANE-BASED COATING MASS WITH A CATALYTIC, OXIDATIVE
AND DEODORIZING EFFECT

This invention relates to a catalytic composition which comprises a coating of a coating material on a support and is obtainable by applying the coating material, comprising (1) a polycondensate of at least one hydrolysable organosilane and also, if desired, one or more compounds of glass-forming elements, and (2) particles of one or more transition metal oxides, the weight ratio of transition metal oxide particles to polycondensate being from 10:1 to 1:10, to the support and subjecting the applied coating material to thermal treatment. The invention also relates to a process for preparing this catalytic composition and to its use for the purpose of deodorizing or oxidizing organic components or carbon.

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	First Named Inventor	Thomas BENTHIEN
	COMPLETE IF KNOWN	
	Application Number	09/937,464
	Filing Date	
	Group Art Unit	
	Examiner Name	

As a below named inventor, I hereby declare that:

My residence, mailing address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

SILANE-BASED COATING MASS WITH A CATALYTIC,
OXIDATIVE AND DEODORIZING EFFECT

(Title of the Invention)

the specification of which

☐ is attached hereto

OR

☒ was filed on (MM/DD/YYYY)

04/05/2000

as United States Application Number or PCT International

(if applicable).

Application Number

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and was amended on (MM/DD/YYYY)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

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				YES	NO
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☐ Additional foreign application numbers are listed on a supplemental priority data sheet PTO/SB/02B attached hereto:

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NAME OF SOLE OR FIRST INVENTOR :

☐ A petition has been filed for this unsigned inventor

Given Name
(first and middle [if any])

Thomas

Family Name
or Surname

BENTHIEN

Inventor's
Signature

Th B

Date

13/02/02

Residence: City

State

Country

Citizenship German

Mailing Address

Rosenstrasse 12, 86899 Landsberg am Lech,

Mailing Address

Germany

DEX

City

State

ZIP

Country

NAME OF SECOND INVENTOR:

☐ A petition has been filed for this unsigned inventor

Given Name
(first and middle [if any])

Stefan

Family Name
or Surname

FABER

Inventor's
Signature

Stefan Faber

Date

18/02/02

Residence: City

State

Country

Citizenship German

Mailing Address

Kraewigstrasse 25, 66687 Wadern,

Mailing Address

Germany

DEX

City

State

ZIP

Country

☐ Additional inventors are being named on the 2 supplemental Additional Inventor(s) sheet(s) PTO/SB/02A attached hereto.

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ADDITIONAL INVENTOR(S) Supplemental Sheet

Page 1 of 2

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventor

Given Name (first and middle [if any])

Family Name or Surname

Gerhard

JONSCHKER

Inventor's
Signature

Date

1/30/02

Residence: City

State

Country

Citizenship German

Mailing Address

Gruenewaldstrasse 12, 66583 Spiesen-

Mailing Address

Elversberg, Germany

DEX

City

State

ZIP

Country

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventor

Given Name (first and middle [if any])

Family Name or Surname

Stefan

SEPEUR

Inventor's
Signature

Date

18/02/02

Residence: City

State

Country

Citizenship German

Mailing Address

Kirchstrasse 22, 66787 Wadgassen-

Mailing Address

Schaffhausen, Germany

DEX

City

State

ZIP

Country

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventor

Given Name (first and middle [if any])

Family Name or Surname

Helmut

SCHMIDT

Inventor's
Signature

Date

Feb. 26, 2002

Residence: City

State

Country

Citizenship German

Mailing Address

Im Koenigsfeld 29, 66130 Saarbruecken-

Mailing Address

Guedingen, Germany

DEX

City

State

ZIP

Country

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PTO/SB/02A (11-00)

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DECLARATION

ADDITIONAL INVENTOR(S)

Supplemental Sheet

Page 2 of 2

Name of Additional Joint Inventor, if any:		<input type="checkbox"/> A petition has been filed for this unsigned inventor	
Given Name (first and middle [if any])		Family Name or Surname	
6-00 <u>Philipp</u>		<u>STOEBEL</u>	
Inventor's Signature <u>Philipp Stoebel</u>		Date <u>02.15.02</u>	
Residence: City	State	Country	Citizenship German
Mailing Address <u>Skalleystrasse 3, 66125 Saarbruecken, DEX</u>			
Mailing Address <u>Germany</u>			
City	State	ZIP	Country
Name of Additional Joint Inventor, if any:		<input type="checkbox"/> A petition has been filed for this unsigned inventor	
Given Name (first and middle [if any])		Family Name or Surname	
Inventor's Signature		Date	
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Mailing Address			
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Name of Additional Joint Inventor, if any:		<input type="checkbox"/> A petition has been filed for this unsigned inventor	
Given Name (first and middle [if any])		Family Name or Surname	
Inventor's Signature		Date	
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Mailing Address			
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PATENTS

Attorney Docket No. 24448-0030

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

First Inventor Name: Thomas BENTHIEN

PCT Int'l. App. No.: PCT/EP00/03020

Int'l. Filing Date: April 5, 2000

For: SILANE-BASED COATING MASS WITH A CATALYTIC,
OXIDATIVE AND DEODORIZING EFFECT

Commissioner for Patents
Washington, D.C. 20231

Sir:

**POWER OF ATTORNEY BY ASSIGNEE
AND STATEMENT UNDER 37 CFR 3.73**

Institut für Neue Materialien gemeinnützige GmbH ("INM"), a German corporation, states that it is the assignee of the entire right, title and interest in the US national phase of the PCT International Application identified above by virtue of an assignment from the inventors, a copy of which is attached hereto.

As assignee of record of the entire interest of the above identified application, INM hereby appoints the practitioners at Customer Number 25213 as its attorneys and agents to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.

The undersigned is authorized to act on behalf of the assignee.

Institut für Neue Materialien gemeinnützige GmbH

By:  
Helmut Schmidt Werner Bonke
Managing Director Business Director

Date: Sep. 9, 2001

093744-0340